UCRL-JC-131214
PREPRINT

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This paper was prepared for submittal to the
30th Boulder Damage Symposium: Annual Symposium on
Optical Materials for High Power Lasers
Boulder, Colorado
September 28 - October 1, 1997

December 23, 1998

Lawrence Livermore National Laboratory

Role of starting material composition in interfacial damage morphology of hafnia silica multilayer coatings

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ABSTRACT

Flat bottom pits, previously shown to be unstable above a critical fluence, may be impacted by interfacial characteristics of the multilayer. TEM cross-sections reveal multilayer coatings deposited from hafnium have fewer interfacial voids than those deposited from hafnia. To correlate this reduction with the occurrence of flat bottom pits, multilayer high reflectors deposited by reactive electron-beam evaporation from both metallic and oxide sources were damage tested with 3-ns, 1064-nm pulses. To shift the electric-field peaks to the adjacent interface, half of the samples included an additional buried half-wave of silica in their all quarter-wave design. All quarter-wave reflectors deposited from hafnia had flat bottom pit damage in the outer six layers at fluences of 20 J/cm^2. Interfacial damage also occurred in hafnium deposited coatings at fluences as low as 20 J/cm^2, but at significantly different depths. The only interfacial damage observed on the all quarter-wave coatings was at the substrate multilayer interface. Flat bottom pits were observed in the buried half-wave coatings with a correlation to electric-field position and preference to interface type.

Keywords: flat bottom pits, coating interfaces, reactive e-beam evaporation, hafnia silica multilayers

1. INTRODUCTION

The construction of the National Ignition Facility (NIF) for use in studying Inertial Confinement Fusion (ICF) requires large aperture optics that withstand high fluences over a reasonable life span.[1] Therefore process improvements in thin film deposition that increase the optic lifetime in high-powered lasers are worth investigating, provided they are economically prudent. It has already been demonstrated that laser systems have some tolerance for damaged optics without adverse power loss, therefore it is not necessary to strive for the complete elimination of damage.[2] However, by minimizing damage that grows catastrophically under repetitive illumination, the optic life span is increased thus lowering operational costs.

Flat bottom pits are interfacial in nature and typically occur within the first few outer layer pairs. Generally the flat bottom pit depth corresponds to the electric field peaks at the hafnia over silica interface, where electric fields are highest.[3] Flat bottom pits in hafnia silica multilayer coatings have been observed to grow upon further illumination so they can impact the optic lifetime.[3] Flat bottom pits are created around nodular ejection sites as well as in the absence of visible defects. Dijon has theorized that the presence of nanoscale absorbing seeds are responsible for the formation of flat bottom pits.[4] This model shows that there is sufficient energy at electric field peaks to create a plasma leading to film buckling.[5-6] Additionally the plasma emission creates localized radiation damage.

During development of hafnium deposition for high damage threshold coatings,[7] it was found that flat bottom pits did not occur in hafnium deposited coatings irradiated at NIF fluences, whereas they did occur in hafnia deposited coatings. A number of factors could explain the starting material compositional dependence on the occurrence of this damage morphology, including elimination of nanoscale absorbing seeds or improvement of interfacial strength. It is not surprising that the interfacial quality in these coatings is material dependent since the hafnia layers are polycrystalline and rough while the silica layers are amorphous and smooth, hence material growth occurs on two very different surfaces. However, there is also a starting material compositional dependence on interfacial quality as illustrated in figure 1. Coatings deposited from hafnium and silica have a noticeable reduction in interfacial voids compared to hafnia and silica deposited coatings. The impact of starting material composition on the presence of absorbing seeds is unknown.

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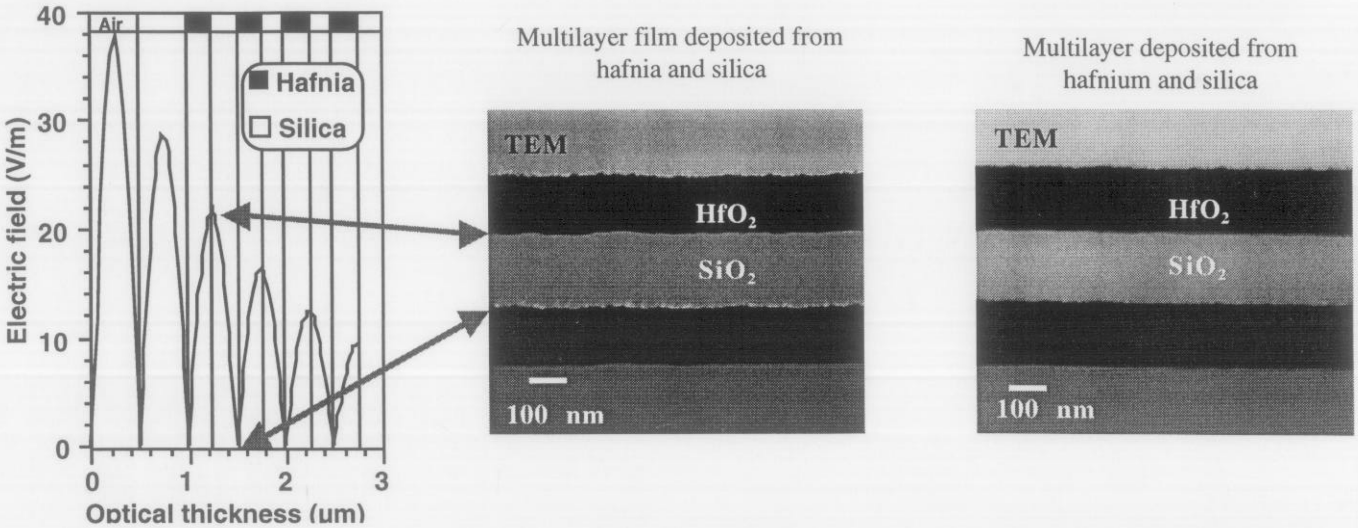

Fig.1 Comparison of interface quality as a function of starting material composition and corresponding electric-field profile.

2. EXPERIMENTAL PROCEDURE

2.1 Coating design

To improve understanding of the role of electric field on both interfaces, a buried half-wave design was developed to shift the electric-field peak in the outer layers from the hafnia grown on silica interface to the silica grown on hafnia interface as illustrated in figure 2. The symmetry of the electric field profile, calculated with Macleod Thin-Film Design Software,[8] within this design provides an opportunity to simultaneously expose the two different interfaces to high electric fields to determine differences in interfacial strength between the two different materials. In addition to shifting the electric-field peak locations, the peak amplitude was also unfortunately significantly increased. Therefore it is anticipated that the damage threshold of the buried half-wave coating will be lower than the all quarter-wave design.

2.2. Mirror coating preparation

Mirrors were evaporated onto 50-mm diameter BK-7 substrates by reactive e-beam deposition in the Spectra-Physics 96" coating chamber used for ICF coatings. The hafnia deposited coatings were from early NIF coating development efforts. Subsequent development efforts were focussed on hafnium evaporation, with an emphasis on achieving an absorptance comparable to standard hafnia deposited films of less than 100 ppm.

The coatings with the modified designs were deposited only from hafnium and silica. A special masking tool was used to enable masking of half of the samples during the deposition of the second half of the buried half wave. With this only exception, the two coating designs were deposited simultaneously under identical conditions.

2.3. Laser testing conditions

Using a Nd:YAG laser with a 1064-nm, 3-ns pulse length at S-polarization, the samples were damaged and then examined for different morphologies. The beam was adjusted for a far field, circular, Gaussian profile with a diameter of 1.1-mm at $1/e^2$ of the maximum intensity.

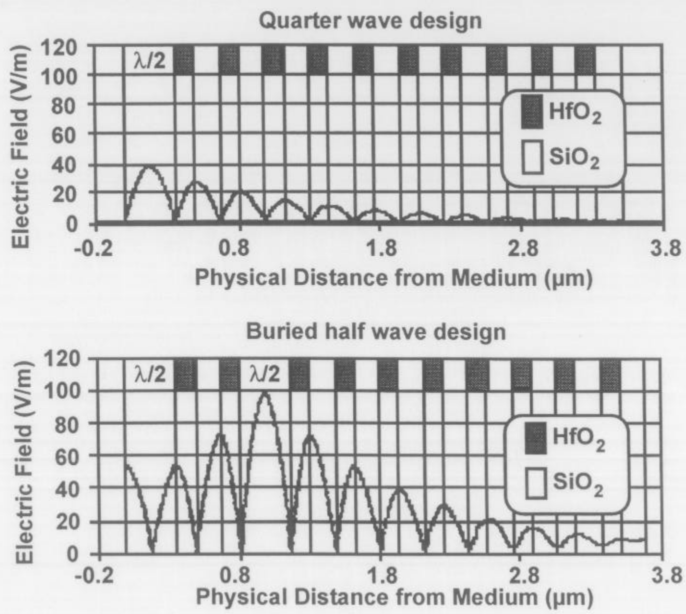

Fig. 2 Electric-field profile amplitude and interface peak locations are different for the two designs.

This profile was used to calculate the peak fluence. Circular areas of the coating were tested a 10° angle of incidence at fluences between 10 and 30 J/cm^2 by raster scanning with a 0.2 mm step as described elsewhere.[9] The raster scan technique was adopted because of the low defect density and the desire to identify the fluence limiting defects over larger areas. However two significant disadvantages of the raster scan technique are the possibility of laser conditioning and modification of damage created at lower fluence during subsequent higher fluence scans.

Tests were conducted on a total of six samples including an equal number of all quarter-wave and buried half-wave coatings. The average area tested per sample was 45.6 cm^2 or approximately 56% of the coated area. In an attempt to duplicate the flat bottom pit damage morphology normally observed in hafnia deposited coatings, one sample of each coating type was tested at single sites at 40 J/cm^2. The testing was performed in previously raster scanned, but undamaged areas. Any damage and its subsequent growth was noted after each shot until it had stabilized.

2.4. Microscopy

Using bright and dark field optical microscopy, the samples were examined before and after testing for detection of any damage larger than 10 μm. Once damage occurred, an Atomic Force Microscope (AFM) was used to characterize the depth of the damage to determine potential sites for Scanning Electron Microscopy (SEM) and Focused Ion Beam (FIB) cross sectioning. The interfacial quality was assessed by Transmission Electron Microscopy (TEM). Cross-sectional TEM specimens were prepared by a technique similar to that described elsewhere.[10] Two pieces of the substrate containing the deposited film were epoxied face-to-face, potted in a 3 mm diameter tube, sliced, lapped, dimpled then low angle ion milled until perforation using single-sided sector ion beam modulation conditions. Specimens were then examined in a JEOL 200CX.

3. RESULTS AND DISCUSSION

3.1. Hafnia starting material

Coatings deposited from hafnia and silica have a significant number of flat bottom pits at various interfacial depths as shown in the FIB micrographs in figure 3. These samples were irradiated at 30 J/cm^2 at 10 ns, which correlates to 20 J/cm^2 at 3 ns assuming a pulse length scaling relation of $\tau^{0.35}$. Similar morphologies are also reported for measurements at 3 ns.[2-3] The

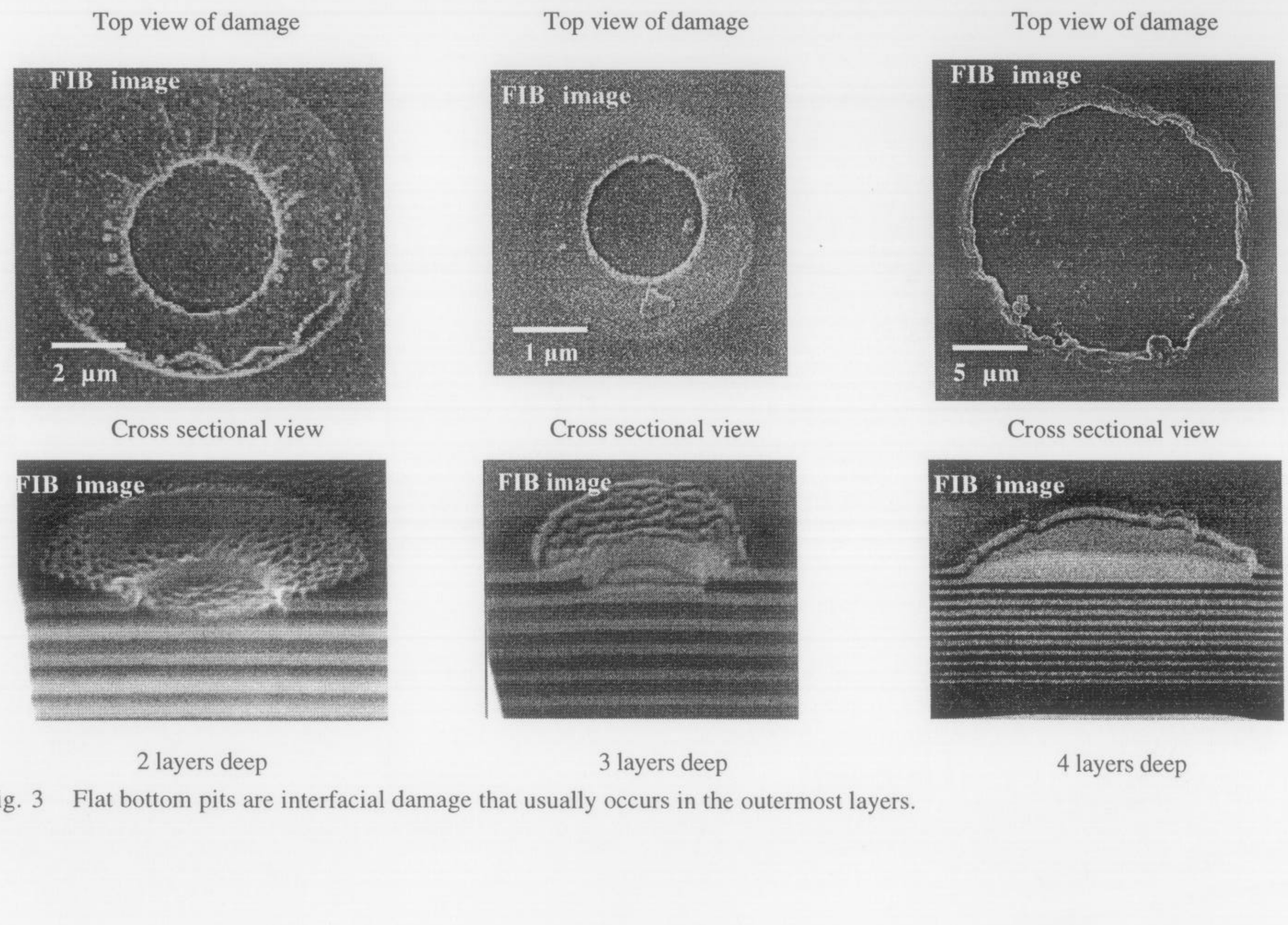

Fig. 3 Flat bottom pits are interfacial damage that usually occurs in the outermost layers.

cross sections reveal the true depth of the flat bottom pits whereas AFM measurements are subject to calibration errors and SEM images yield only subjective depth information.

Typically the damage is one to three layer pairs deep corresponding to the interfaces at electric field peaks. However, there are exceptions such as the three layer deep pit in figure 3. This is particularly troublesome to explain by the theory of nanoscale absorbing seeds at high electric field locations since the damaged interface is at an electric field minimum. The two layer deep flat bottom pit shows evidence of plasma interaction throughout the entire pit by temperature-induced surface roughening typical of plasma scalds. However the bottom of the three and four layer deep flat bottom pits are smooth indicating a mechanical failure of the damaged layer possibly after the plasma extinguishes when the film rapidly goes into tension during cooling. This delamination mechanism has also been observed in the outer layer of coatings with thin overcoats.[11] Although the damage may be initiated by nanoscale absorbing seeds at high electric field locations, the interfacial quality and plasma presence may also play critical roles in the ultimate flat bottom pit depth.

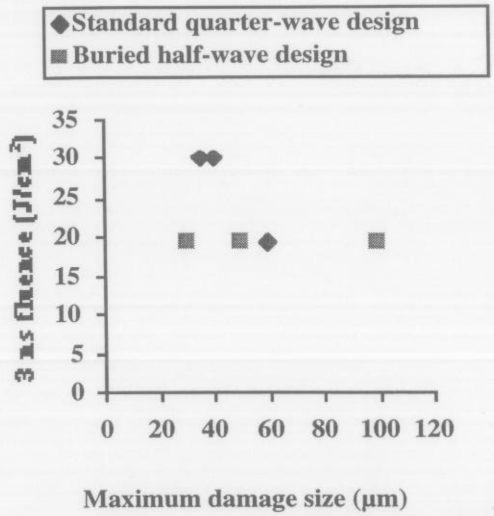

Fig. 4 Damage in all quarter-wave coating was smaller at higher fluences than the buried half-wave coating.

3.2. Hafnium starting material

As illustrated in figure 4, the buried half-wave coatings typically have larger damage sites for a given fluence and catastropic sites at a lower fluence than the all quarter-wave coatings. For the buried-half wave samples, most were raster-scanned at increasing fluences until approximately 20 J/cm^2 before testing was stopped due to damage. The all quarter-wave coatings

Top view of damage morphology. FIB cross section of damage site reveals substrate interfacial damage. Multilayer interfacial damage likely due to nonuniform adhesion between defect and coating.

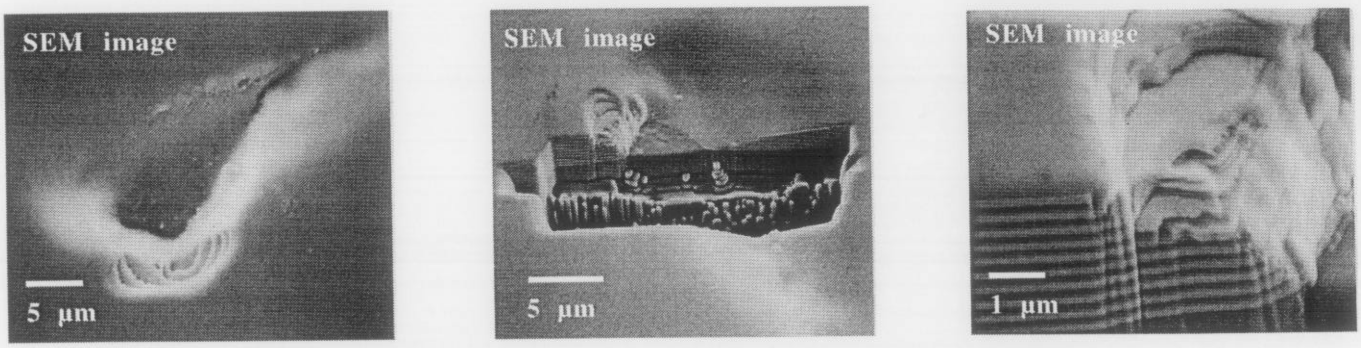

Fig. 5 Damage morphology of site 1 of all quarter-wave reflector has substrate interfacial damage.

Top view of damage morphology FIB cross section reveals substrate damage

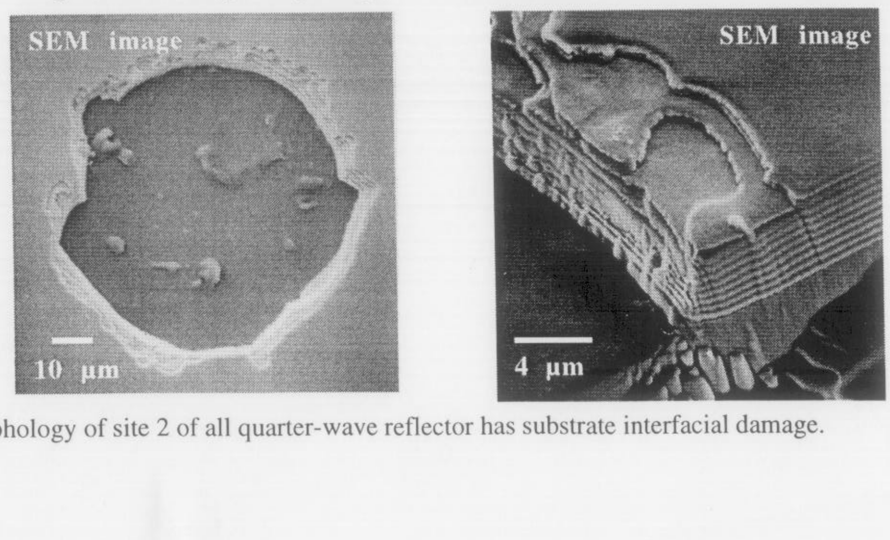

Fig. 6 Damage morphology of site 2 of all quarter-wave reflector has substrate interfacial damage.

were raster scanned at increasing fluences until approximately 30 J/cm^2 before testing was stopped. It is not surprising that the buried half-wave coatings had a lower damage threshold because the maximum electric field peak is more than twice that in the all quarter-wave design.

The interfacial damage that occurred in the all quarter-wave design is shown in figures 5 and 6. In each case the damage occurs at the substrate multilayer interface as revealed in the FIB cross sections. Previous cross sections have revealed that critical nodular defects are at or near the substrate interface, so these damage sites may have been of nodular origin. The damage illustrated in figure 6 was first noted at approximately 15 J/cm^2, but unfortunately was not examined for depth. This allows for two possible explanations for the damage morphology. First, the original formation of the pit could have been shallower, making the damage a result of the electric field that then grew to its final depth. However, if the pit stayed at its original depth of about 3.5 µm, the electric field peak should not be a factor in the damage, therefore a different mechanism may be responsible for multilayer substrate interfacial damage. Other damage morphologies were also found, mainly plasma scalding and nodular ejection that are stable at fluences up to 30 J/cm^2.

The interfacial damage in the buried half-wave design, as illustrated in figures 7 and 8, are very different than the all quarter-wave design and look more like typical flat bottom pits with the exception of the pit depth. In these damage sites the pit was six to ten layers deep and appeared to be initiated at a nodular seed. In contrast, hafnia deposited all quarter-wave design coatings typically have flat bottom pit damage that is much shallower at two to six layers deep.

Inspection of the electric-field profile, illustrated in figure 9, confirms that the damage depth correlated to interfaces at electric field peaks. Although there were electric field peaks of similar magnitude closer to the surface, flat bottom pits did not occur at these depths. This result suggests a difference in the strength of the two types of interfaces with the silica grown on hafnia

Top view of damage morphology | FIB cross section reveals damage likely originated from a nodular ejection | Interfacial damage is 6, 8, & 10 layers deep

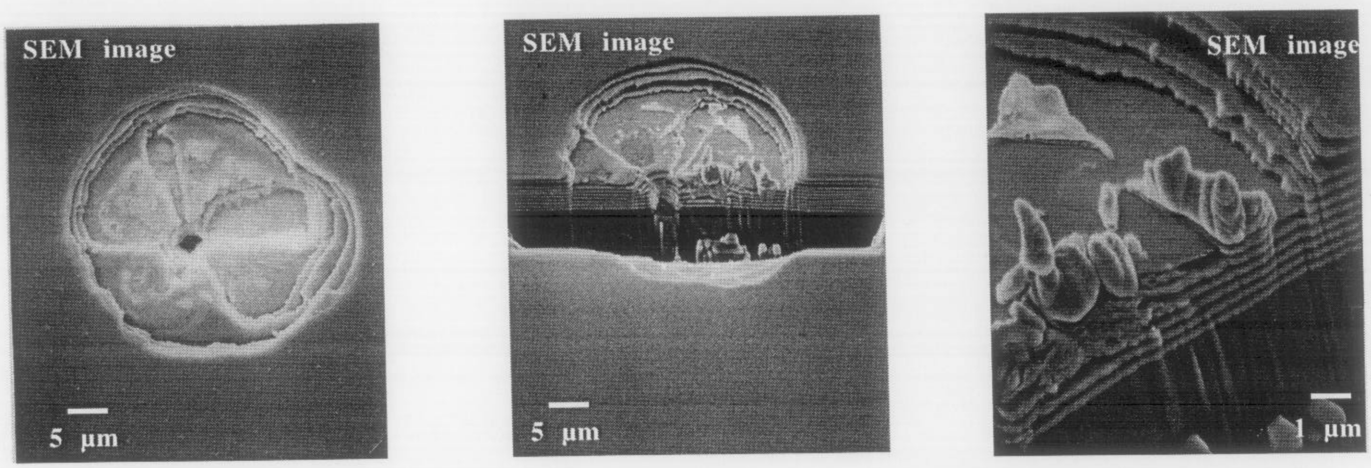

Fig. 7 Damage morphology of site 1 of buried half-wave design has multilayer interfacial damage

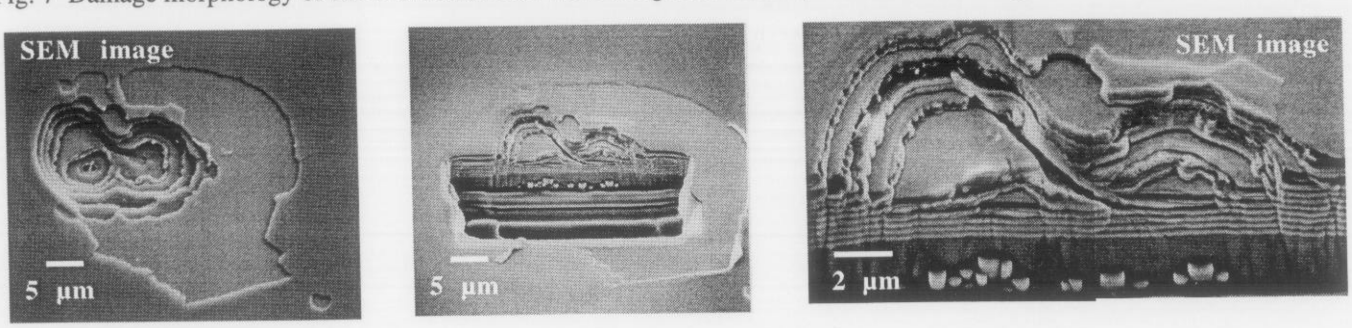

Fig. 8 Damage morphology of site 2 of buried half-wave design has multilayer interfacial damage.

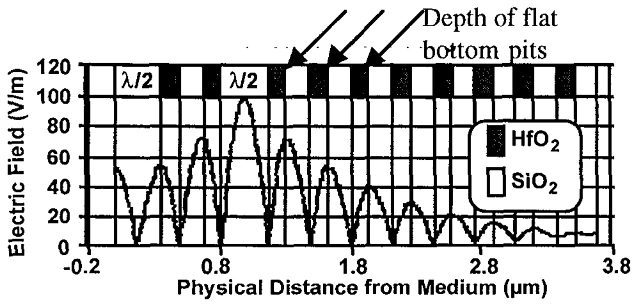

Fig. 9 Flat bottom pits correlate to the hafnia over silica interfaces with electric field peaks.

being stronger. Interestingly, for the hafnia deposited coatings, the opposite interface appears be stronger based on the amount of interfacial voiding illustrated in figure 1. If this experiment would be repeated with hafnia deposited coatings, they would likely have shallow flat bottom pits with depths one or three layers corresponding to the electric field peaks, but at the opposite interface of the hafnium deposited coatings.

After the N-on-1 testing at 40 J/cm^2 (the maximum capacity of the testing laser) the all quarter-wave coating did not show any damage, whereas the buried half-wave coating damaged by means of scalding and nodular ejection at three out of four tested areas.

4. CONCLUSIONS

The samples with coatings that were deposited from a metallic hafnium source showed a significant reduction in the number of flat bottom pits when compared with the coatings deposited from an oxide source. The improved coating interfaces that result from using a metallic as opposed to oxide source can therefore be correlated with a lower occurrence of flat bottom pits and a higher damage threshold. The damage depth of the buried half-coating design illustrated preferential damage at the hafnia grown over silica interface for equal electric field intensities suggesting weaker interfacial strength.

5. ACKNOWLEDGEMENTS

The authors would like to acknowledge the LLNL microscopy support of Jim Hughes (AFM), Jim Yoshiyama (SEM), and Mark Wall (TEM). Farid Ghafghaichi at Accurel Systems did the FIB cross sectioning and imaging. The coatings were generated by Alfred Nini and Roy Cardozo at Spectra-Physics. This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under contract No. W-7405-Eng-48.

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